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THE STUDY OF ALKALI HALIDE CRYSTAL COUNTERS

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A STUDY OF ALKALI HALIDE CRYSTAL COUNTERS

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Purpose

The object of this work was to study the behavior of electrons and holes in alkali halides. It was desired to measure the mobility of electrons and holes, and also to study their production, their trapping, and the temperature dependence of these processes. For reasons which are not understood, the results have been largely negative, although several interesting phenomena have been noticed which deserve further investigation.

Method

The experimental method was similar to that used by McKay[1][2] in his experiments on diamond, but with certain improvements in the electronic instruments. The samples studied were in the form of single crystals and were used in the circuit shown in Figure 1. In most cases the crystal was a few tenths of a millimeter thick and about one centimeter square. The electrodes were usually semi-transparent evaporated layers of aluminum (aluminum was chosen because of its small stopping power for cathode rays and alpha particles), although in the early experiments graphite was used. The capacitance C in Figure 1 consists of the capacitance of the crystal, amplifier input circuit, and associated wiring.

The crystals were normally good insulators, so that no current flowed through the resistor R. However, if ionizing particles (cathode rays or alpha particles in this case) entered the crystal,

free electrons and holes were produced and moved (presumably) under the influence of the electric field. In these experiments, short (10^{-8} second) pulses of 10 KV cathode rays, or, in some cases single 5 MEV alpha particles, entered the crystal through one of the electrodes. The cathode rays or alpha particles stopped in a short distance, producing electrons and holes. Then, depending on the polarity of the electric field, either the electrons or the holes drifted toward the opposite electrode. This produced a voltage pulse across R, which was amplified and displayed on the screen of an oscilloscope. Since the current lasted only as long as there were free electrons (or holes) in the crystal, the shape of the pulse on the oscilloscope screen gave the time taken for the charges to drift from one end of the crystal to the other.

Let this time be t, the thickness of the crystal d, and the velocity of drift v_d.

Then

$$v_d = \frac{d}{t} \qquad \text{(I)}$$

and the mobility is by definition given by

$$v_d = \mu E \qquad \text{(II)}$$

where E is the electric field. We will now consider the shape of the pulse in detail.

Free electrons and holes will move about in a Brownian motion, upon which is superimposed their drift in the electric field, until they either recombine, reach an electrode, or get trapped at some

crystal imperfection. We will neglect any direct recombination. For very small electric fields almost all of the charges will be trapped before they can get to an electrode, while for sufficiently large electric fields the drift velocity will be large enough so that they will almost all reach the electrodes. As long as the carriers do not reach electrodes, there will be a characteristic time T for trapping, such that if N_o electrons (or holes) are produced at time zero, then the number remaining at time t will be

$$N = N_o e^{-\frac{t}{T}} \qquad \text{(III)}$$

T is given by

$$T = \frac{1}{\rho \sigma v} \qquad \text{(IV)}$$

where ρ is the density of traps, σ their cross section, and v the velocity of the electrons in their Brownian motion.

Since the time constant RC, Figure 1, is large compared with the other times involved, we will consider the pulse from the standpoint of the charge appearing on the capacitor C. If a charge q moves in the crystal a distance dx in the direction of the electric field, then the charge dQ induced on C will be[3]

$$dQ = q \frac{dx}{d} \qquad \text{(V)}$$

The signal voltage seen by the amplifier will be

$$V = \frac{1}{C} \int dq \qquad \text{(VI)}$$

Since in this case all the charge carriers are produced at one end of the crystal, only electrons or only holes, depending on the polarity of the electric field, will give an appreciable contribution to Q. One can then be sure of working with only one type of carrier at a time, which is one of the advantages of this method.

If we put

$$dx = v_d\, dt = \mu E dt \qquad \text{(VII)}$$

then

$$V(t) = \frac{\mu E}{Cd} \int_{t=0}^{t} q\,dt \qquad \text{(VIII)}$$

If $q = Ne$, where N is the number of free electrons (or holes), e is the charge of an electron, and $N = N_o e^{-\frac{t}{T}}$, then

$$V(t) = \frac{\mu E}{Cd}\, eN_o \int_{t=0}^{t} e^{-t/T}\, dt = \frac{\mu E T e N_o}{Cd}\,(1 - e^{-t/T}) \qquad \text{(IX)}$$

This of course is true only if t is small enough so that the electrons do not reach an electrode: i.e. $\mu E t < d$. For longer times

$$V = \frac{\mu E}{Cd}\, eN_o \int_{t=0}^{d/\mu E} e^{-t/T}\, dt = \frac{\mu E T e N_o}{Cd}\,(1 - e^{-d/\mu E T}). \qquad \text{(X)}$$

This is independent of t, since by this time all the electrons have either been trapped or have reached an electrode. The quantity $\omega\tau = \mu E T$ is the "Schubweg".

We then write

$$V(t,\omega\tau) = \frac{\omega\tau\, eN_0}{Cd}\left(1 - e^{-t/T}\right) \text{ for } t \leq \frac{d}{\mu E} . \qquad \text{(XI)}$$

$$V(\omega\tau) = V(\infty,\omega\tau) = \frac{\omega\tau\, eN_0}{Cd}\left(1 - e^{-d/\omega\tau}\right) \text{ for } t \geq \frac{d}{\mu E} . \qquad \text{(XII)}$$

The function $V(\omega\tau)$ is plotted in Figure 2. As the electric field in the crystal increases there is a "saturation" of the pulse height, corresponding to having nearly all the electrons reach the opposite electrode. The work of Flechsig[4][5] on photocurrents in NaCl indicates that saturation can be reached with crystals of thickness of the order of 0.1 mm. Flechsig's data gives $\mu T = 5 \times 10^{-7}$ cm^2/volt at room temperature.

By measuring the final pulse height $V(\omega\tau)$ as a function of E one obtains the product μT[1.][6.], and by determining the shape of the pulse one obtains T. In this manner μ is obtained.

The shape of the pulse is characterized by the "rise time", which is defined as the time it takes for the pulse to rise from 10 % to 90 % of its final value. From equation XI it is easily shown that for $\omega\tau \ll d$, the rise time is

$$t_{10\text{-}90} = T \ln 9 = 2.2T \qquad \text{(XIII)}$$

In general, for any $\omega\tau$, using equations XI and XII:

$$t_{10-90} = T \ln \frac{9e^{1/v(\omega\tau)} + 1}{e^{1/v(\omega\tau)} + 9} \qquad \text{(XIV)}$$

Recently, Witt[7] has reported that NaCl crystals will count alpha particles. Witt obtained very small pulses, and no indication of saturation, as the pulse heights were closely proportional to the electric field across the crystal.

One of the difficulties with this type of experiment is "polarization". After current has passed through the crystal there are electrons trapped near the anode and holes trapped near the cathode. These charges create an internal space charge field which tends to neutralize the applied field. Thus, the pulse height decreases with time as more and more carriers get trapped. With some materials the polarization can be removed by turning off the applied field and allowing the space charge to be neutralized with the aid of light or cathode rays which free the trapped charges.

Apparatus

1. Electronics

A block diagram of the electronic equipment (excluding that associated with the electron gun and the vacuum gauges) is shown in Figure 3. The preamplifier was designed for low noise, high impedance input, and low impedance (200 ohm) output. Its circuit is shown in Figure 4. In order to minimize stray capacitance,

the preamplifier was mounted inside the vacuum chamber, close to the crystal holder. For heat dissipation, the tubes were supported by being waxed with Apiezon W into holes in a piece of 1/16 inch thick aluminum. The 0.57 μμfd capacitor shown in Figure 4 enables calibration of the circuit directly in terms of charge appearing on the grid of the first tube by applying a step function signal of known voltage through this capacitor.

The overall sensitivity of the amplifiers is such that about 8.5×10^{-15} coulombs on the grid of the first preamplifier tube gives a deflection of one centimeter on the oscilloscope screen, at full gain. This of course depends to some extent on the capacitance of the crystal in the holder. The waveform response is such that a step function pulse at the input of the preamplifier gives a pulse on the oscilloscope screen with a rise time of about 10^{-8} second. This is in good agreement with what would be expected from the calculated characteristics of the preamplifier and the advertised characteristics of the Hewlett-Packard amplifiers and the Tektronix oscilloscope. The noise in the amplifiers corresponds to about three millimeters deflection on the oscilloscope.

The peak reading vacuum tube voltmeter is used for pulse height measurements, since this is more convenient and more sensitive than measuring the pulse height directly on the oscilloscope screen. The scalar is used as an extra visual and audio indication of correct functioning, and also, by use of a pulse height discriminator, to detect pulses which might be barely above the noise level.

2. Experiment Chamber

A cross section sketch of the vacuum chamber is shown in Figure 5. The chamber has three removable ports, four inches in diameter; one is used for the electron gun, and the others for quartz windows through which the crystal can be illuminated. One of the ports also has a vacuum seal with a rotatable shaft which enables a piece of polonium foil to be brought in front of the crystal in order to test for alpha particle counting. The top and bottom plates are also removable. On the top plate are attached the crystal holder, preamplifier, and electrical terminals. There is a tube on the bottom plate which may be dipped in liquid nitrogen so that vapors may be trapped before they can condense on the crystal or the high voltage insulators.

The crystal holder is shown in Figure 6. The electron beam enters the crystal from the left through a hole in the holder. The holder is surrounded by a radiation shield which is not shown in the drawing.

All of the experiments were carried out in a vacuum of 10^{-5} mm Hg or better.

3. Electron Gun

The electron gun was taken from a DuMont type 3 GP1 cathode ray tube and modified by placing the original oxide coated cathode with a filament of 0.005 inch thoriated tungsten. This is necessary in order to avoid "poisoning" of the cathode when air is let into the system between experiments. The modification was done by Mr. Allen B. Wilson of the Electrical Engineering Research Laboratory.

The gun is used with about 1400 volts between the filament and second anode, and ten kilovolts between filament and ground. The focus, intensity, and positioning circuits are conventional for this type of tube, the filament current can be varied by means of a Variac in the primary of the filament transformer. The maximum beam current is about one microampere. The beam is pulsed by applying to the grid of the gun a positive pulse of about 50 volts and about 5×10^{-9} second duration.

4. Pulse Generator

The basic pulse generator circuit is shown in Figure 7. The Western Electric 275C relay has contacts which are wet with mercury and in a high pressure hydrogen atmosphere in order to allow very fast make and break. The relay is used without any modification. It operates sixty times per second.

The pulse generator gave pulses with rise time less than 7×10^{-9} second, which is the rise time of the oscilloscope amplifier. It was used for testing and calibrating the apparatus, as well as to trigger the electron gun and the oscilloscope sweep.

A similar, but more refined, pulse generator has been described in the literature by Garwin.[6]

Results

In general, the conclusions which may be drawn from the experiments are qualitative. In many cases the results could not be repeated very well at different times and with different crystals. Possible reasons for this are:

(1) Polarization. Because of the poor reproducibility, one could not be very sure that the crystals were completely unpolarized. A dependable method for depolarizing them was not found.

(2) Formation of traps. Bombardment of a crystal with cathode rays and ultraviolet light for extended times might produce vacancies and/or coagulations of vacancies which could trap electrons and holes.

(3) Filling and emptying of traps. Freed electrons and holes could fill traps and thus change the schubweg. These traps might then empty out thermally, giving an additional time dependence of the schubweg.

(4) Ionic conductivity. Appreciable ionic conductivity could contribute to polarization and formation of traps when the electric field is left on for sufficient time. It is thought that the conductivity is not high enough at room temperature and below to do much in the few minutes' time that the field was usually left on.

(5) Cold work and annealing. Repeated handling, cooling, and warming of a crystal could change its state of cold work and thus change the schubweg. In a few cases crystals were annealed and then tried. Although this data is very meager, no differences were found between annealed and unannealed specimens.

(6) Surface effects. Since the cathode rays penetrate only about 10^{-4} cm., minute cracks and dislocations might influence the number of electrons and holes which can

get into the body of the crystal. Future experiments probably should be done with higher energy cathode rays.

Because of the poor reproducibility, the apparatus was checked frequently with either no crystal in the holder or with a diamond in the holder. Diamond was used because the author has had considerable experience with it[9] and data on polarization, pulse heights, etc. can be reproduced very well with it. From these checks it was concluded that the effects reported below are ascribable to the crystals and not to the apparatus.

Results with Alpha Particles

The following alkali halides were tested for alpha particle counting: KI, NaI, LiF, KCl, NaCl, and KBr. None of them gave detectable pulses except NaI. A description of the results with NaI follows.

Two crystals, about 0.35 mm thick, were cleaved from the same large crystal. The faces were coated with "Dag" dispersion no. 154 (colloidal graphite in alcohol). At liquid nitrogen temperature both crystals counted individual alpha particles, with about 1000 volts applied. The pulses were of about equal size for either polarity of applied electric field, indicating that electrons and holes could be made to move about equally. The pulse heights were such that it takes at most 200 e.v. of energy to produce one free electron-hole pair. At room temperature one of the crystals did not seem to count at all. The other counted by means of electron conduction about as well as it did at the low

temperature, but counted somewhat more weakly by means of hole conduction.

There was no indication of any gamma ray counting from a 6 mc. radium source held about five inches from the crystal.

Results with Cathode Rays

In most cases the cathode ray pulses were of about 10^{-14} coulomb and lasted for 10^{-8} second. This was measured by allowing the pulsed beam to fall on the crystal holder with nothing between the electrodes. This figure does not take into account elastic reflection of the incident electrons. However, by putting positive potentials of up to 5000 volts on the crystal holder and noting the increase in pulse size, it was determined that inelastically reflected electrons were no more than about twenty percent of the total.

Sodium Iodide. Three specimens of NaI were tested for response to cathode ray pulses. Their thicknesses were 0.25 mm., 0.45 mm., and 0.32 mm., respectively. The first had graphite electrodes, while the other two had evaporated aluminum electrodes. Experiments were done at room temperature and, with liquid nitrogen coolant, at about -165° C, as measured by a thermocouple attached to the crystal holder.

At both temperatures there were conduction pulses with either direction of electric field, indicating that both electrons and holes are mobile. At the low temperature the largest pulses were for electron conduction, while at room temperature the largest were for hole conduction. (This is in disagreement with the results for

alpha particle counting. Since the alphas penetrate about ten times as far as the cathode rays, it may be that this disagreement is due to different temperature dependence of surface effects for electrons and holes.) In all cases the pulses were small--of the order of magnitude of one electron or hole for each incident electron. Electron pulses were about the same at both temperatures. At low temperature the hole pulses were about one-fourth as high as the electron pulses, while at room temperatures they were about four times as high as the electron pulses.

The pulse heights seemed to be independent of the electric field, at least for voltages across the crystal of the order of 300 or greater. The rise time of the pulses was in all cases less than or equal to 10^{-8} second (the rise time of the apparatus).

These results seem to indicate that the free electrons and holes cannot move farther than about 3×10^{-5} cm. in the crystal. For if one assumes that each incident 10 KV electron produces about one thousand free pairs, then from the size of the pulse these secondaries must move only a thousandth of the thickness of the crystal before being trapped. (This is less than the penetration depth of the cathode rays!)

Since this occurs in a time of 10^{-8} second or less, the drift velocity can be as small as 3×10^{3} cm/sec, which, for an electric field of 10^{4} volt/cm. would mean a mobility of about 0.3 cm^2/volt sec. Since the "saturation" field, the time, and the number of carriers might be considerably overestimated, the value of the mobility might be considerably greater than indicated above.

The NaI showed fairly definite polarization effects. If a crystal had been counting cathode rays by means of electrons for some time and the applied field was then turned off, there were pulses in the opposite direction, gradually decreasing in size, for several seconds, indicating a space charge field. The analogous situation occurred if the crystal was originally counting by means of holes. It appeared that the crystal could be depolarized by light from a Hanovia high pressure mercury arc, or an infrared heat lamp (of the type sold in drug stores), or by a steady cathode ray beam. There was some evidence for a greater ($\sim$50 %) schubweg when the crystal was illuminated by either lamp) than when in the dark. There were no detectable ($> 5 \times 10^{-10}$ ampere) steady photocurrents with either of the light sources.

Potassium Chloride. Six specimens of KCl, with thicknesses from about 0.2 mm. to 0.4 mm. were tested. They all had evaporated aluminum electrodes. The behavior was very erratic, and one of the specimens gave no response at all. Several of the crystals had been annealed at 600° C; they were then soft enough so that they could easily be bent once through a right angle, after which they became work hardened and brittle again. No differences were noticed between the annealed and unannealed crystals.

The crystals gave small electron pulses at both temperatures. Hole pulses were even smaller and more erratic, at room temperature; there did not seem to be any hole pulses at the low temperature.

Some polarization effects were noticed. The material could be depolarized by the mercury arc (but not very dependably) or by a steady cathode ray beam.

Potassium Bromide. Five crystals of KBr were used. They were cut from the same large crystal and had evaporated aluminum electrodes. Their thicknesses ranged from 0.38 to 0.86 mm. Of these, two (the thinnest and the thickest) gave fairly reproducible results and two behaved more erratically. The other one gave very little response compared to the others, and was not studied thoroughly. No hole motion was ever detected; the following results apply to electron motion.

Maxiumum pulse sizes were large; in most cases five to ten times as large as for the other materials, but in one case eighty times as large. In addition, it was found that under some conditions the rise times were long, and the mercury light had a very pronounced effect on both the size and shape of the pulses. Experiments were done at room temperature and at liquid nitrogen temperature. No significant differences were found, except that at room temperature the behavior was more erratic.

Pulses were comparatively large, as mentioned above, and the rise times were long--of the order of 0.2 microsecond. There was polarization, as evidenced by decreasing pulse height after a few seconds. Depolarization could be effected by light from the mercury lamp, with the applied field turned off. It was necessary to illuminate for a minute or so to get the most depolarization, but this was not very dependable. No pulse height versus applied field measurements could be taken because of the depolarization difficulties, but it appeared that larger pulses resulted from higher fields at least up to 4500 volts across the crystal thickness.

If light from the mercury lamp was shone on the crystal while the field was on, the pulse height would increase by a factor ranging from 1.5 to 10, and simultaneously the rise time would decrease to 10^{-8} second? This took about ten seconds, and one could watch on the oscilloscope screen the pulses gradually getting larger and sharper. There was little or no change of pulse height with time in this state. The light could be removed, and the crystal would remain in this state. It could be put back into its previous, long rise time, state by turning off the field and illuminating with the light for a minute or so. Insufficient illumination here would leave a crystal in a state such that the pulses would rise part way in a very short time, and then slowly the rest of the way. The transition back and forth between the two states could be repeated indefinitely. If, while in the short rise time state, the applied field was turned off, the pulse height would not decrease to zero immediately, but would decrease to about one-half its original value (in the same direction) immediately, and then slowly ($\sim$30 seconds) to zero. In the long rise time state it would go to zero immediately. These things could all be repeated also with smaller (about one-fifth) initial cathode ray pulses.

If the light was passed through a filter cutting out wavelengths below about 5300Å (Corning 3-69), it would still take the crystal from the slow to the fast rise time state, but not as quickly and maybe not as completely as the light from the bare mercury arc. However, the light of wavelength above 5300Å would not take the crystal from the fast to the slow rise time state.

Photocurrents of about 5×10^{-9} ampere could be obtained with light from the bare mercury arc. When the light passed through filters cutting off at 4200A or 5300A the current was about halved. A filter cutting off at 6400A cut the photocurrent to about one tenth its maximum value. A piece of pyrex glass 2mm. thick cut the current to about two-thirds its maximum value. From this it appears that the most effective light is the green line, 5461A. This is within the F band of KBr. Although no change of photocurrent with time was noticed, possibly because the galvanometer used has a period of five seconds, there was polarization; if one allowed photocurrent to flow, then covered the light and turned off the field, then uncovered the light, there was current in the opposite direction--indicating a space charge field in the crystal.

The above results seem to be consistent with the following model. From the pulse heights, the schubweg must be about one two-hundredth the thickness of the crystal, or about 3×10^{-4} cm. If now photocurrent is passed through the crystal, it becomes polarized. We now assume that in this polarized state most of the potential drop is in the vicinity of the cathode, where the primary electrons enter. This corresponds to a positive space charge in the crystal. If this region of high field is several times larger than the original schubweg, the new schubweg will be this distance because of the extremely high electric field, and the pulse height will be larger. Also, the rise time of the pulses will be many times smaller, since the high field will mean a larger drift velocity.

A possible explanation for the lack of hole pulses may be the following. KBr was the only material tested which gave photocurrents, and it is likely that these currents were due to F centers. The work of Dutton[10] indicates that holes in KBr may be captured and annihilated by F centers. It may be that this is what happens to the holes in this experiment.

One can make the same rough calculation for mobility in KBr as was made above for NaI. Assume that the electrons (in the long rise time state) travel 3×10^{-4} cm. in 10^{-7} second with a field of 10^5 volt/cm. The mobility is then 3×10^{-2} cm^2/sec.-volt. This of course should be regarded as a lower limit.

Footnotes

(1) McKay: Phys. Rev., 74, 1606 (1948).

(2) McKay: Phys. Rev., 77, 816 (1950).

(3) This can be derived very easily by conservation of energy.

(4) Flechsig: Zeitz. f. Phys., 46, 788 (1928).

(5) Flechsig: Phys. Zeits., 32, 843 (1931).

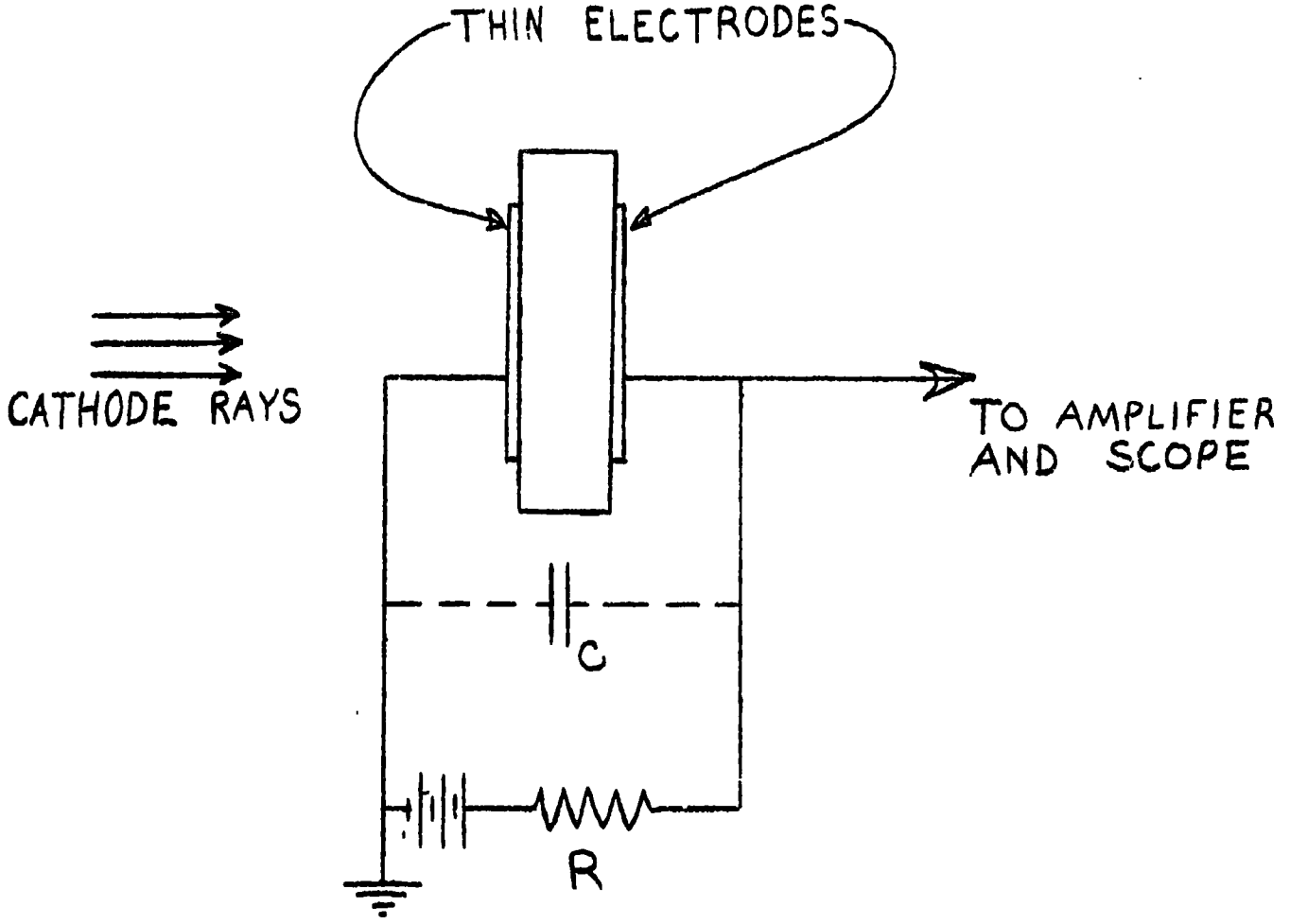
(6) See also Mott and Gurney, Electronic Processes in Ionic Crystals, p. 122.

(7) Witt: Zeits. f. Phys. 128, 442 (1950).

(8) Garwin: Rev. Sci. Inst., 21, 903 (1950).

(9) Final Report, Navy Contract N7-onr-303, T.O. 5 Part II (1950), Carnegie Institute of Technology.

(10) D. B. Dutton. Thesis, University of Illinois, 1952.



BASIC CIRCUIT FOR CRYSTAL

FIGURE 1

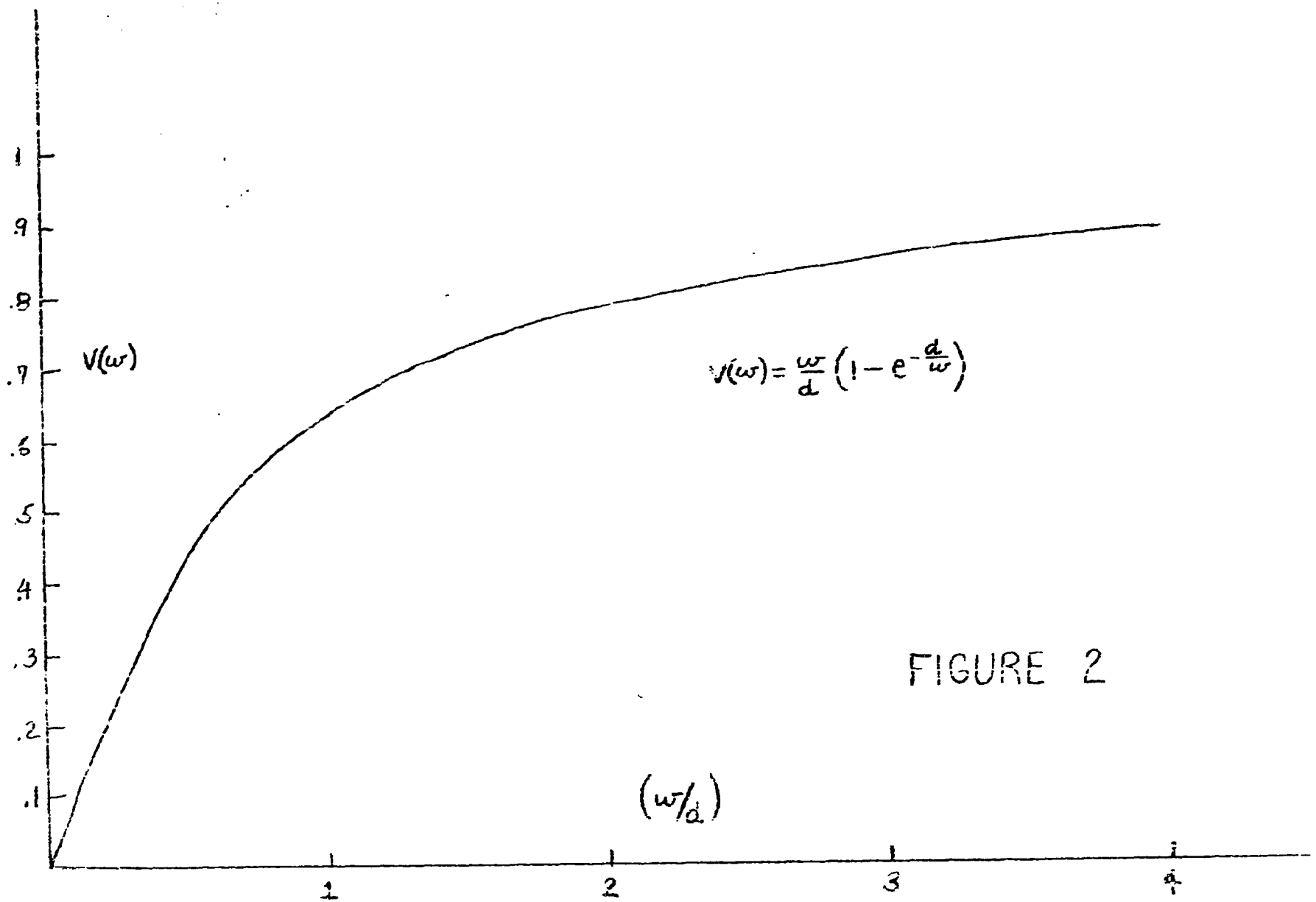
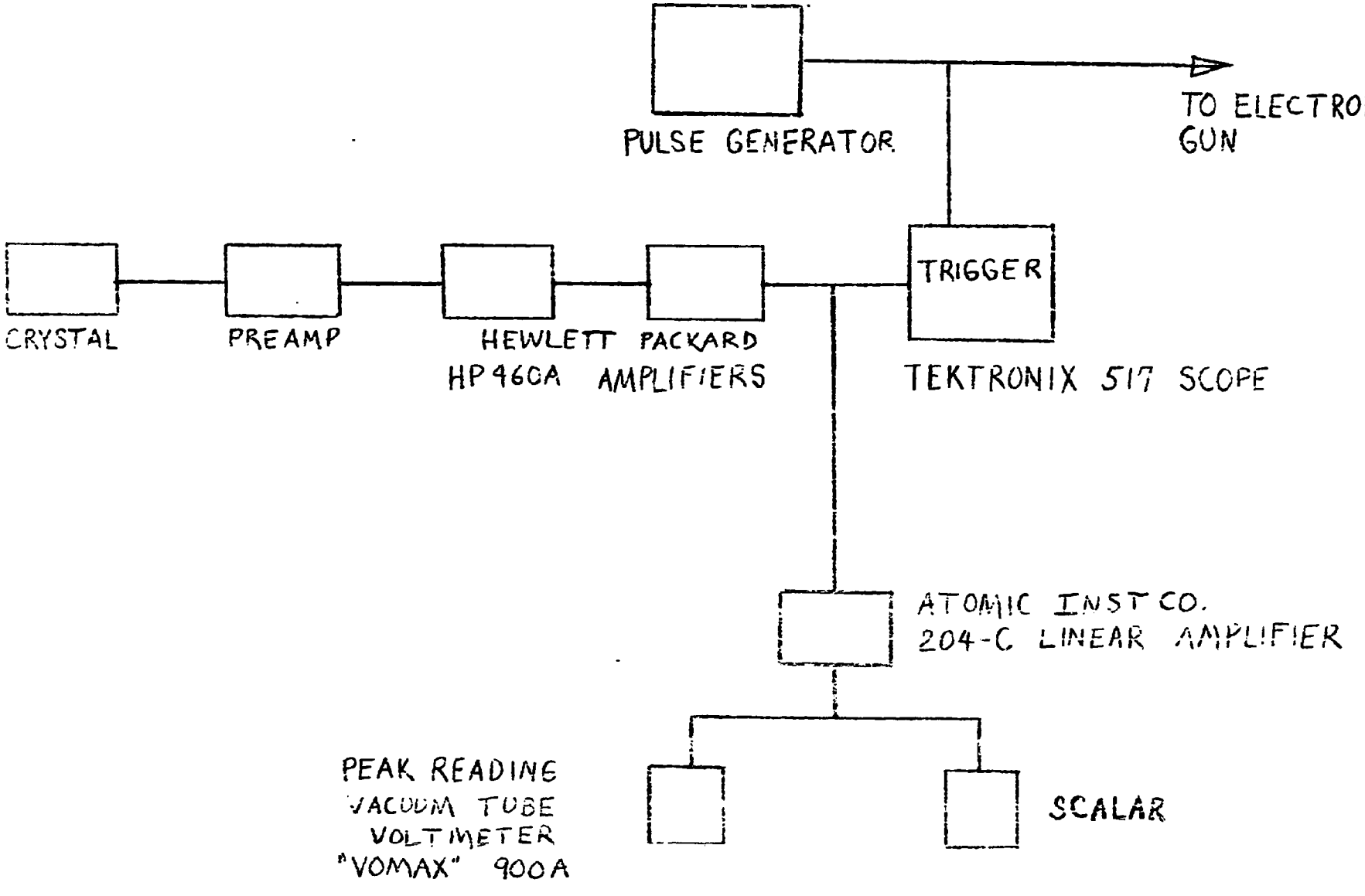
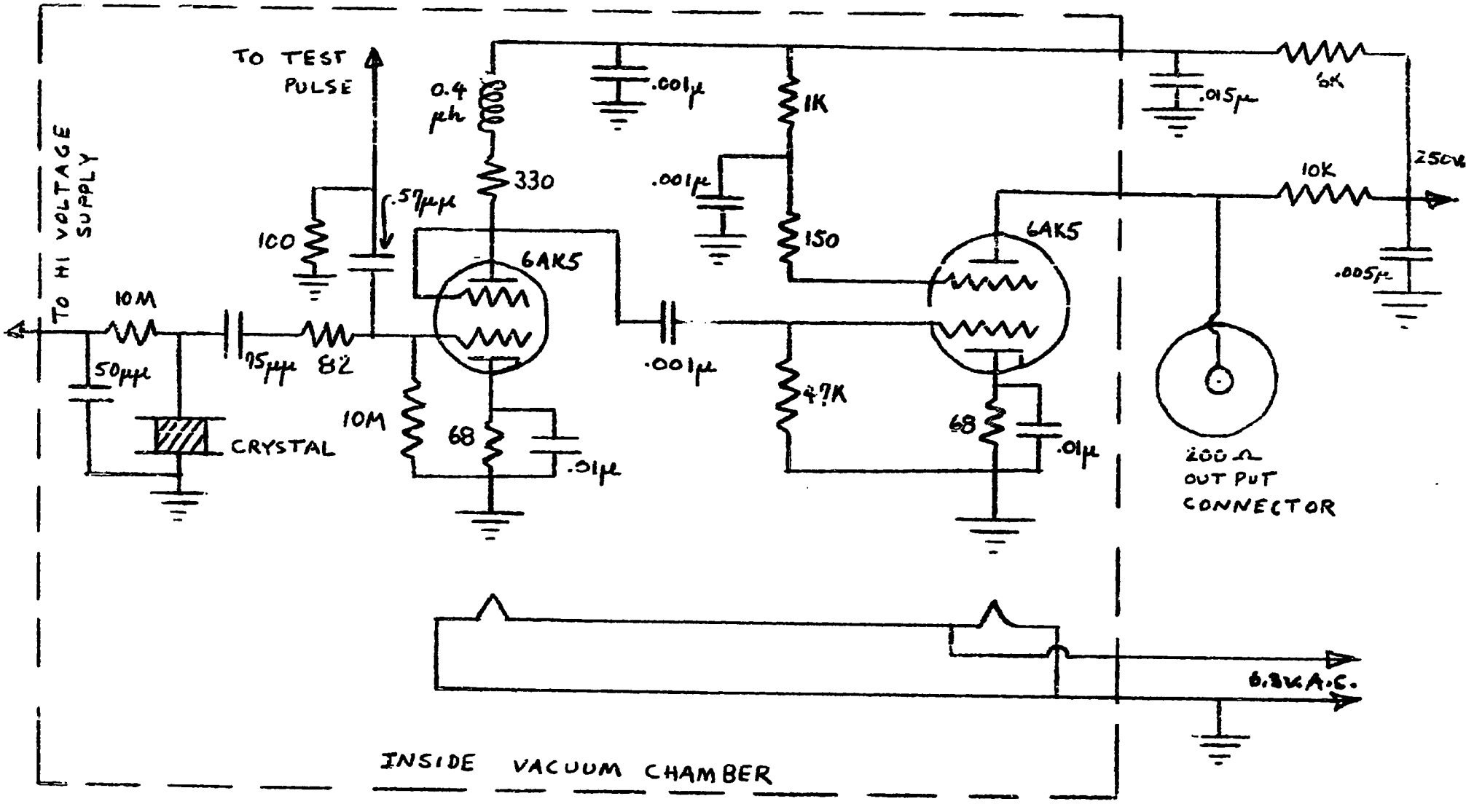

1
.9
.8
V(ω)
.7
.6
.5
.4
.3
.2
.1
$V(\omega) = \frac{\omega}{d}\left(1 - e^{-\frac{d}{\omega}}\right)$
(ω/d)
1
2
3
4

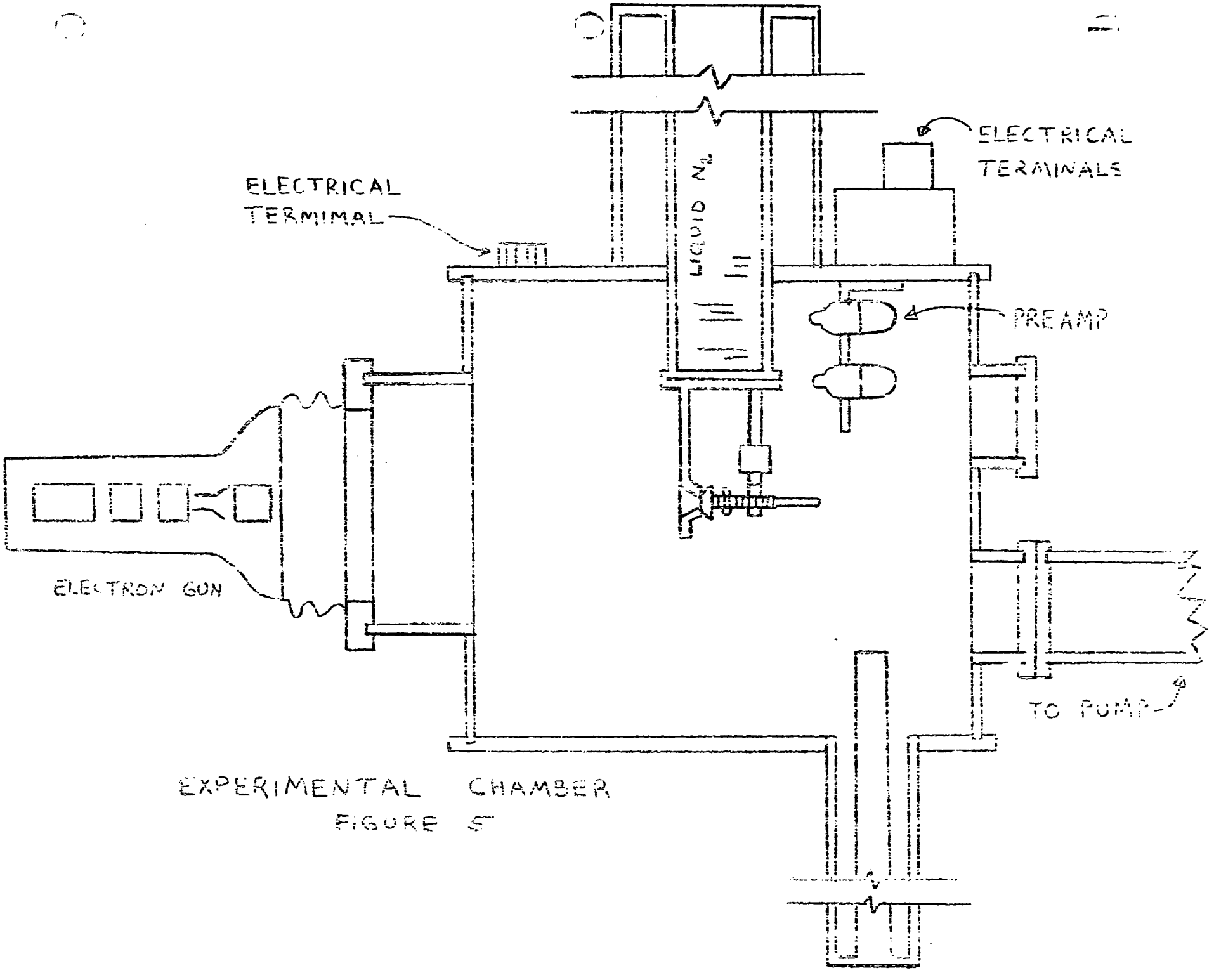
FIGURE 2



BLOCK DIAGRAM – FIGURE 3

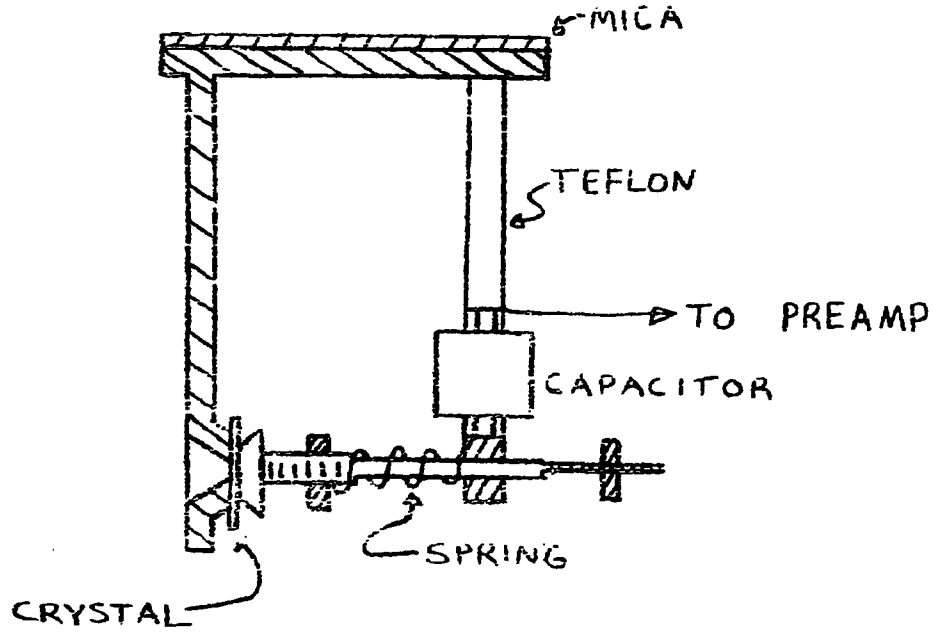


PREAMPLIFIER FIGURE 4



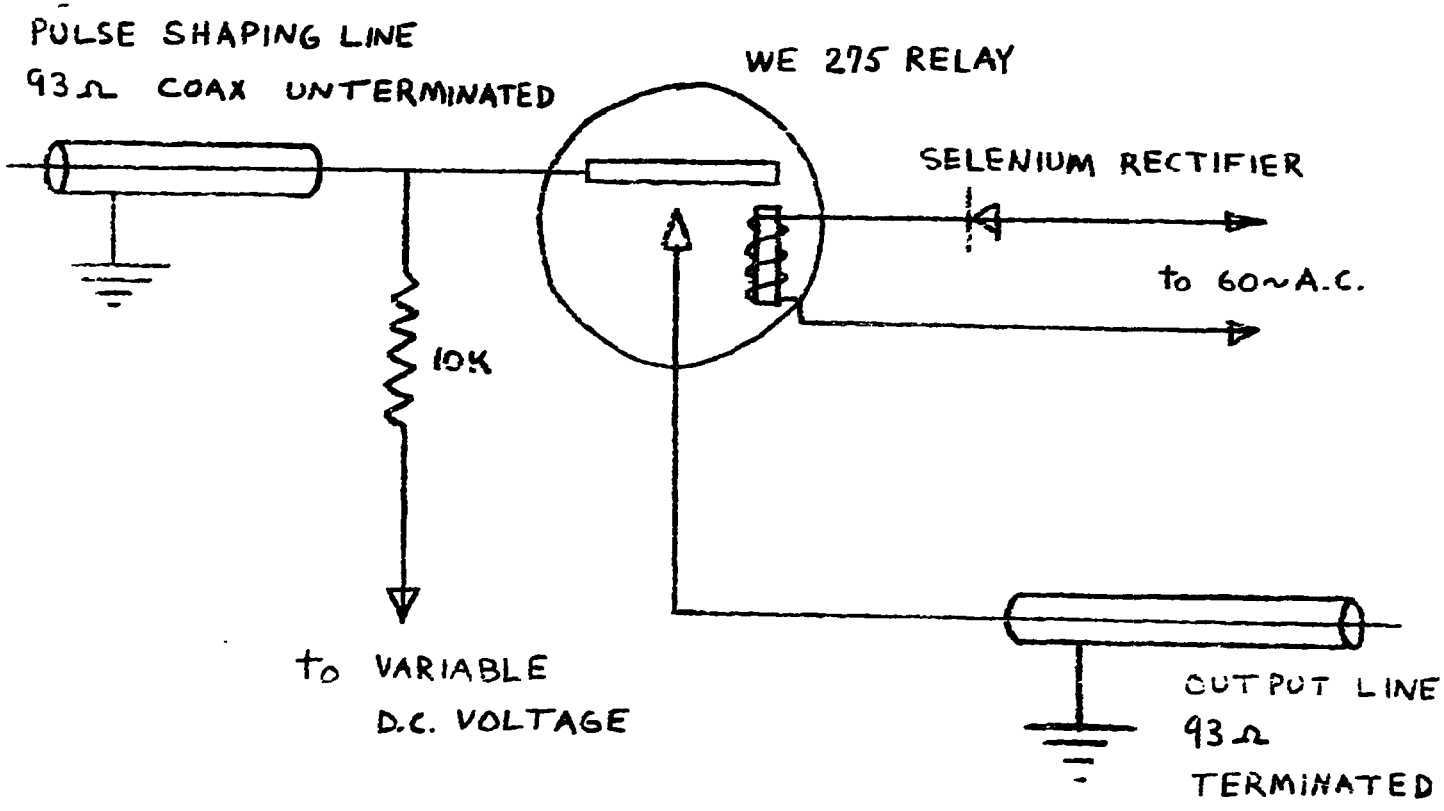
EXPERIMENTAL CHAMBER

FIGURE 5



CRYSTAL HOLDER

FIGURE 6



BASIC PULSE GENERATOR CIRCUIT

FIGURE 7